

# On-demand production of entangled photon pairs via quantum-dot biexciton control

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(Dated: February 16, 2012)

A pulsed laser scenario, designed to prepare an appropriately chosen self-assembled quantum dot with 100% population in a biexciton state, is proposed. The ensuing radiative emission would provide a near perfect (99.5%) high rate, on-demand, source of entangled photons.

*Introduction* – Entangled photon generation is of considerable significance in quantum cryptography where the appearance of unentangled photons in a signal indicates possible eavesdropping. In such cases extremely high-quality entangled photon generation (essentially 100% of the emitted photons being entangled) is required [1]. Other uses of entangled photons would require less entangled-photon purity (e.g. 99%) and include a number of applications such as two-photon absorption [2], microscopy [3], lithography [4] and two-photon coherent tomography [5]. Several of these applications, such as entangled-photon imaging [6] and entangled photon lithography [7], have not been successful in setups where the entangled photons are generated by standard parametric down conversion, since these applications require a high rate of photon-pair generation. Parametric down conversion is very ineffective in this respect: photon pairs are generated in this technique with very small probability. Hence, there is considerable interest in developing new, efficient sources of entangled photons that can prove useful for assorted technical and fundamental applications.

In this paper we propose a nearly-deterministic method for producing entangled photons at a high rate. It resolves numerous problems associated with the recent proposal [8] and demonstration [9] that the decay of quantum-dot systems from the biexciton state to the ground state can be used as a triggered source of entangled photon pairs. The quantum dot decays spontaneously via two exciton states, representing two decay channels. If the exciton states cannot be spectrally resolved (i.e., when the splitting between the exciton states due to anisotropic electron-hole exchange is much less than the radiative linewidths of these states), then welcher-weg information on the two photons is erased. Thus, the generated photons are entangled in polarization basis.

To date, this method has generated photon pairs that are very far from perfectly correlated, due to background noise: The best fidelity of producing the entangled pairs has been poor, basically limited to  $\sim 70\%$  even after

reducing the noise [10]. Noting that one of the main sources of the background noise in these experiments is the radiative decay of the exciton states, we here propose a method that avoids such decay. Specifically, we show that it is possible to achieve *complete* population transfer from the ground state to the biexciton state. In that case, any two consecutive photons that are generated from the quantum dots would be entangled. The first photon is generated when the quantum dot decays from the biexciton to one of the exciton states and the next photon is generated from the decay of the same exciton state to the ground state. We also show that Auger effects, which could add unentangled photon contamination, can be controlled by suppressing recapture of excited electrons or holes using an electric field.

*Model system* – Consider a self-assembled quantum dot with multiple exciton states. For each pair of exciton states there exists a biexciton state, corresponding to both these states being populated. When subject to a linearly polarized electromagnetic pulse with field  $\vec{E}(t)$ , the ground state  $|0\rangle$  is coupled to the exciton states  $|1^+\rangle$ ,  $|2^+\rangle$ ,  $\dots$  and  $|1^-\rangle$ ,  $|2^-\rangle$ ,  $\dots$  by the  $\sigma_+$  and  $\sigma_-$  components of the input pulse, respectively. The same components couple the exciton states to the biexciton states  $|1^+, 1^-\rangle$ ,  $|1^+, 2^-\rangle$ ,  $|1^+, 2^+\rangle$ ,  $|2^+, 2^-\rangle$ , etc. with selection rules depicted in Fig. 1. The frequency of the exciton-to-biexciton transition is offset by the binding energy  $\Delta$  from the corresponding ground-to-exciton transition frequency. In a typical quantum dot,  $\Delta \ll \omega_k$ , where  $\omega_k$  is the transition frequency between states  $|0\rangle \leftrightarrow |k^\pm\rangle$ .

It is important to note that nonradiative transitions (notably Auger recombination) of the quantum dots from the biexciton to the exciton states serve to reduce the probability of obtaining a pair of photons that could otherwise be entangled. Thus, it is necessary to identify systems in which the exciton state left behind after Auger recombination will not radiate. In a typical self-assembled quantum dot, the band gap energy is much larger than the confinement barrier so that the final state of Auger recombination is a highly excited exciton state with the electron or hole in the continuum of the barrier

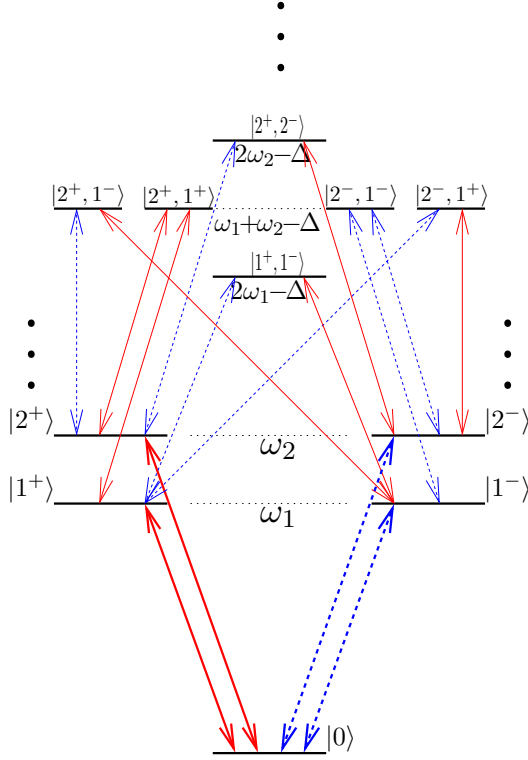


FIG. 1. Level structure and radiative transitions of a quantum dot. Only the first two exciton levels and the corresponding biexciton levels are relevant in this simulation. The red (solid) arrows denote transitions of the  $\sigma_+$  polarized component, while the blue (dotted) arrows denote transitions of the  $\sigma_-$  polarized component of the electric field. Thick lines correspond to ground-to-exciton transitions, while thin lines correspond to exciton-to-biexciton transitions.  $\omega_1$  and  $\omega_2$  are the energy levels of the first and second exciton levels respectively, and  $\Delta$  is the biexciton binding energy.

material. Recapture of such an excited exciton into the quantum dot can be suppressed by sweeping the electron-hole pair out of the system with a dc electric field. It has been demonstrated that with such an electric field, even weakly excited electrons can be tunneled out of a quantum dot in time scales faster than phonon-assisted relaxation [11]. In addition, intra-band relaxation of quasi-free carriers in the conduction or valence band is non-radiative because of energy-momentum conservation constraints [12].

*Fast and efficient population transfer* – To reduce the noise during generation of the entangled photon pairs, we propose a method of complete efficient and rapid population transfer from the ground state  $|0\rangle$  to the lowest biexciton state  $|1^+, 1^-\rangle$ , and computationally demonstrate its efficiency. The exciton states  $|1^+\rangle$  and  $|1^-\rangle$  are coupled to the ground state  $|0\rangle$  via the  $\sigma_+$  and  $\sigma_-$  polarized components of the input pulse, respectively, and from there to the biexciton state  $|1^+, 1^-\rangle$  via the  $\sigma_-$  and  $\sigma_+$  polarization components, respectively.

Since we want to minimize the time spent in the single exciton states  $|1^+\rangle$  and  $|1^-\rangle$  so as to reduce non-entangled photon emission, the total duration  $T_{\text{tot}}$  of the pulse must be much shorter than the radiative decay time  $T_{\text{tot}} \ll \gamma^{-1} \sim 1\text{ns}$ , where  $\gamma$  is the radiative decay rate. For such short times, the time energy uncertainty prevents us from distinguishing between the first exciton states  $|1^\pm\rangle$  and the other, higher states  $|2^\pm\rangle \dots |n^\pm\rangle$ , where  $\omega_n - \omega_1 \sim T_{\text{tot}}^{-1}$ . Thus any computational demonstration of the photon-pair generation must include these higher exciton levels and their biexciton counterparts.

The Hamiltonian in the dipole approximation can be written as

$$\begin{aligned}
 H = & \sum_{k,s} \hbar\omega_k |k^s\rangle \langle k^s| + \sum_k (2\hbar\omega_k - \Delta) |k^+, k^-\rangle \langle k^+, k^-| \\
 & + \sum_{k>j,s,s'} (\hbar\omega_j + \hbar\omega_k - \Delta) |k^s, j^{s'}\rangle \langle k^s, j^{s'}| \\
 & - \sum_{k,s} d_k E^s(t) |k^s\rangle \langle 0| + \text{H.C.} \\
 & - \sum_{k,s} d_{kk} E^s(t) |k^+, k^-\rangle \langle k^-s| + \text{H.C.} \\
 & - \sum_{k \neq j, ss'} d_{jk} E^s(t) |k^s, j^{s'}\rangle \langle j^{s'}| + \text{H.C.}
 \end{aligned} \tag{1}$$

where  $s, s'$  are  $+$  or  $-$  ( $-s$  denotes the sign opposite to  $s$ ),  $E^+$  and  $E^-$  are the  $\sigma_+$  and  $\sigma_-$  polarized components of the electric field  $\vec{E}(t)$  of the input pulse,  $d_k$  is the transition dipole moment between the ground level  $|0\rangle$  and the exciton levels  $|k^s\rangle$  and  $|k^-s\rangle$ , and  $d_{jk}$  is the transition dipole moment between the exciton states  $|j^\pm\rangle$  and the biexciton state  $|j^\pm, k^s\rangle$ . We note that  $|j^s, k^{s'}\rangle$  and  $|k^{s'}, j^s\rangle$  represent identical states.

The calculations rely on the following conditions, appropriate for a typical lens-shaped InGaAs/GaAs self-assembled quantum dot: (i) We assume that the quantum dot is initially prepared in the ground state  $|0\rangle$  with a typical transition frequency of  $\hbar\omega_1 = 1.3\text{ eV}$  [13, 14]. (ii) We limit ourselves to one additional exciton level  $|2^\pm\rangle$ , that is  $\sim 40\text{ meV}$  above the exciton ground state [14]. The biexciton binding energy is taken as a typical value of  $\Delta = 4\text{ meV}$  for all biexciton states. (iii) The ground-to-exciton transitions (with dipole moments  $d_k$ ) in most quantum dots are an s-s transition to the first exciton state, and a p-p transition to the second exciton state. Hence, to first approximation, the transition dipole moment  $d_2$  to the second exciton state  $|2^\pm\rangle$  is  $\sqrt{2}$  times stronger than the transition dipole moment  $d_1$  to the first exciton state  $|1^\pm\rangle$ . (iv) Calculations that treat the electron-hole correlation accurately show that the biexciton-to-exciton radiative decay is  $\sim 1.6$  to  $1.8$  times faster than the radiative decay of the exciton in the size range of interest [15]. When degeneracy (two

channels for emission) is accounted for, this corresponds to a ratio of 0.8 to 0.9 for oscillator strengths. For the proof-of-concept we chose a constant oscillator strength ratio of 0.8 between all the exciton-to-biexciton transitions and the corresponding ground-to-exciton transitions (i.e.,  $d_{jk} = \sqrt{0.8}d_k$ ).

The field  $\vec{E}(t)$  is chosen to be linearly polarized, such that  $E^+(t) = E^-(t)$  and two pulses  $E(t) = E_1(t) + E_2(t)$  are applied: a pulse  $E_1(t)$  with frequency  $\omega_1$  resonant with the  $|0\rangle \leftrightarrow |1^\pm\rangle$  transition, and another pulse  $E_2(t)$  with frequency  $\omega_1 - \Delta$  resonant with the  $|1^\pm\rangle \leftrightarrow |1^+, 1^-\rangle$  transition. Each pulse has a Gaussian profile. The duration of the pulses  $\tau$  should be long enough to keep the second exciton state  $|2^\pm\rangle$  out of the time-energy uncertainty regime:  $\tau \gtrsim 2\pi\hbar/40 \text{ meV} \approx 100 \text{ fs}$ . Additionally, because the ground-to-exciton transition and the exciton-to-biexciton transition have different dipole amplitudes, it is important to access each one separately with a different amplitude pulse. Hence, to keep them out of the time-energy uncertainty regime, we must also demand that the pulse times be larger than  $\tau \gtrsim 2\pi\hbar/4 \text{ meV} \approx 1000 \text{ fs}$ .

On the other hand, the transfer needs to be completed well within the radiative decay time of the exciton levels  $\gamma^{-1} \approx 1 \text{ ns}$ , in order to prevent decay from occurring *during* the transfer process while the population is in a single exciton state, as this will generate unentangled photons.

*Analysis* – The scenario described above is here analyzed for two distinct pulse schemes.

(a) *Sequential-pulse scheme*, where the “ground-to-exciton” pulse  $E_1(t)$  *precedes* the “exciton-to-biexciton” pulse  $E_2(t)$ . In this scheme, all the population is first transferred from the ground state  $|0\rangle$  to the single exciton states  $|1^+\rangle$  and  $|1^-\rangle$ . The population is then excited from these single exciton states to the biexciton state  $|1^+, 1^-\rangle$ . The advantage of such a scheme is that the two pulses  $E_1(t)$  and  $E_2(t)$  hardly interfere [16] insofar as they have little overlap in time. However, the disadvantage is that the system is in a single exciton state between pulses, where it can decay by emitting a single, non-entangled photon, thereby reducing the fidelity of the generated entangled photons. Nevertheless we find that using 1000 fs pulses [Fig. 2], 99.7% of the population is in the biexciton state  $|1^+, 1^-\rangle$ ,  $\sim 0.1\%$  in the ground state  $|0\rangle$ . Only  $\sim 0.2\%$  in one of the other states, which can result in non-entangled photons. The population spends a total of 1.5 ps in each single exciton state  $|1^\pm\rangle$ , resulting in  $\sim 3 \text{ ps} \times \gamma \approx 0.3\%$  additional non-entangled photons.

(b) *Concurrent pulses scheme*. Here the “ground-to-exciton” pulse  $E_1(t)$  and “exciton-to-biexciton” pulse  $E_2(t)$  overlap, transferring the population simultaneously from ground  $|0\rangle$  to the exciton states  $|1^+\rangle$  and  $|1^-\rangle$ , and from the exciton states to the biexciton state  $|1^+, 1^-\rangle$ . The advantage of such a scheme is that the population remains only a minimal amount of time in the single exciton state, thus minimizing the probability of non-entangled photon decay. The disadvantage is that due to interfer-

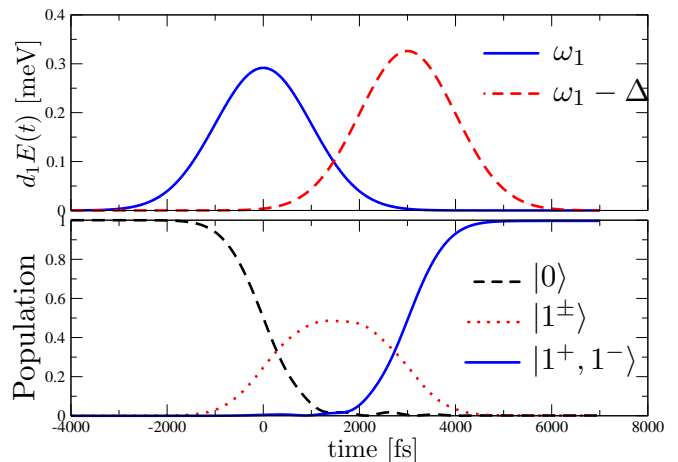


FIG. 2. Simulation results for a scheme with two sequential Gaussian Rabi pulses of width 1000fs. The first pulse of frequency  $\omega_1$ , in resonance with the “ground-to-exciton”  $|0\rangle \leftrightarrow |1^\pm\rangle$  transition, transfers the population from the ground to the single exciton states. The following pulse of frequency  $\omega_1 - \Delta$ , in resonance with the “exciton-to-biexciton”  $|1^\pm\rangle \leftrightarrow |1^+, 1^-\rangle$  transition, transfers the population from the single exciton states to the biexciton state. The result of this scheme leaves 99.7% of the population in the biexciton state and  $\sim 0.1\%$  of the population in the ground state, resulting in at most only  $\sim 0.2\%$  non-entangled (“bad”) photons generated. During the transfer the population spends a total of 3ps in the single exciton states.

ence effects between the pulses, the populations oscillate in time more dramatically than in the sequential-pulse scheme, resulting in the (slightly) lower fidelity [Fig. 3]. The best result found for the population using 1000 fs pulses is 99.1% in the biexciton state  $|1^+, 1^-\rangle$  and  $\sim 0.5\%$  in the ground state  $|0\rangle$  which leaves  $\sim 0.4\%$  in one of the other states, which can result in non-entangled photons. The population spends a total of 0.35 ps in each single exciton state  $|1^\pm\rangle$ , resulting in  $0.7 \text{ ps} \times \gamma \approx 0.07\%$  additional non-entangled photons.

Hence, rather unexpectedly, the sequential-pulses scheme performs better, despite the longer time spent in the single exciton state. However, for systems with a larger radiative decay rate  $\gamma$ , e.g. a system with  $\gamma^{-1} \approx 0.1 \text{ ns}$ , the concurrent-pulses scheme might be better since the added decay probability from a single-exciton state in the sequential-pulses scheme will be larger, negating the advantage of this scheme over the concurrent-pulses scheme.

*Summary* – In conclusion, we propose a highly effective population transfer technique to avoid radiative decay of exciton states in a quantum dot and thus dramatically increase the fidelity of generating entangled photon pairs by spontaneous decay of the biexciton state. When the quantum dot is prepared by appropriate pulses in the biexciton states, with no initial population in the exciton states, contamination due to exciton decay can be

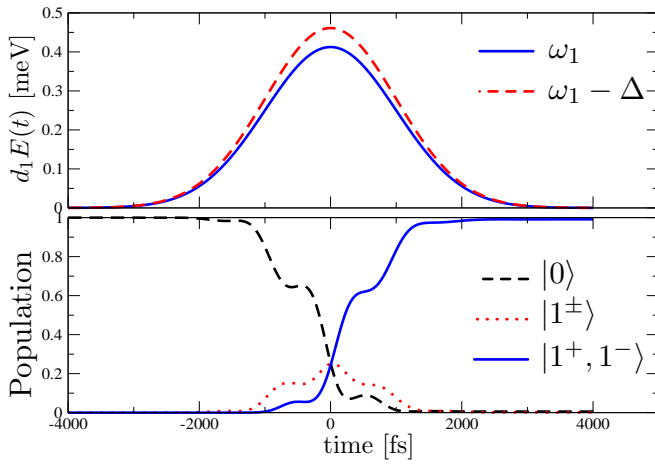


FIG. 3. Simulation results for a scheme with two concurrent Gaussian Rabi pulses of width 1000fs. One pulse is of frequency  $\omega_1$ , in resonance with the “ground-to-exciton”  $|0\rangle \leftrightarrow |1^\pm\rangle$  transition, and the other pulse is of frequency  $\omega_1 - \Delta$ , in resonance with the “exciton-to-biexciton”  $|1^\pm\rangle \leftrightarrow |1^+, 1^-\rangle$  transition. Both together transfer the population from the ground state, through the single exciton states, to the biexciton state. The result of this scheme leaves 99.1% of the population in the biexciton state and  $\sim 0.5\%$  of the population in the ground state, resulting in at most only  $\sim 0.4\%$  non-entangled (“bad”) photons generated. During the transfer the population spends a total of 0.7ps in the single exciton states. Note the oscillatory nature of the population transfer, compared with the sequential transfer in Fig. 2, a feature that is due to the interference inherent in such concurrent pulses.

avoided. Any two consecutive photons that would be emitted due to cascade decay from the biexciton state would then be entangled in the  $(\sigma_\pm)$  polarization basis. We showed, using two laser pulses, how to prepare the quantum dot in the biexciton state and that 99.8% entangled photon generation is achievable using realistic system parameters. Higher fidelity of transfer is more challenging but not impossible, and is best carried out using optimal control techniques in conjunction with the specific experimental arrangement that is adopted.

Finally, we noted that in a quantum dot, the probability of nonradiative decay from biexciton to exciton states due to Auger effects is not completely eliminated, but that this can be overcome by choosing dots where the resulting high-energy exciton is above the confinement energy, whereby it can be removed from the system, as discussed above.

The realistic requirements of the proposed scheme make it a viable method for nearly-deterministic, high-rate, on-demand production of entangled photon pairs, opening the possibility of a variety of quantum applications [2–7].

*Acknowledgement* PB and SD thank Professor G. D. Scholes for various discussions during the early stages of this work. Support from the Natural Sciences and En-

gineering Research Council of Canada is gratefully acknowledged.

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